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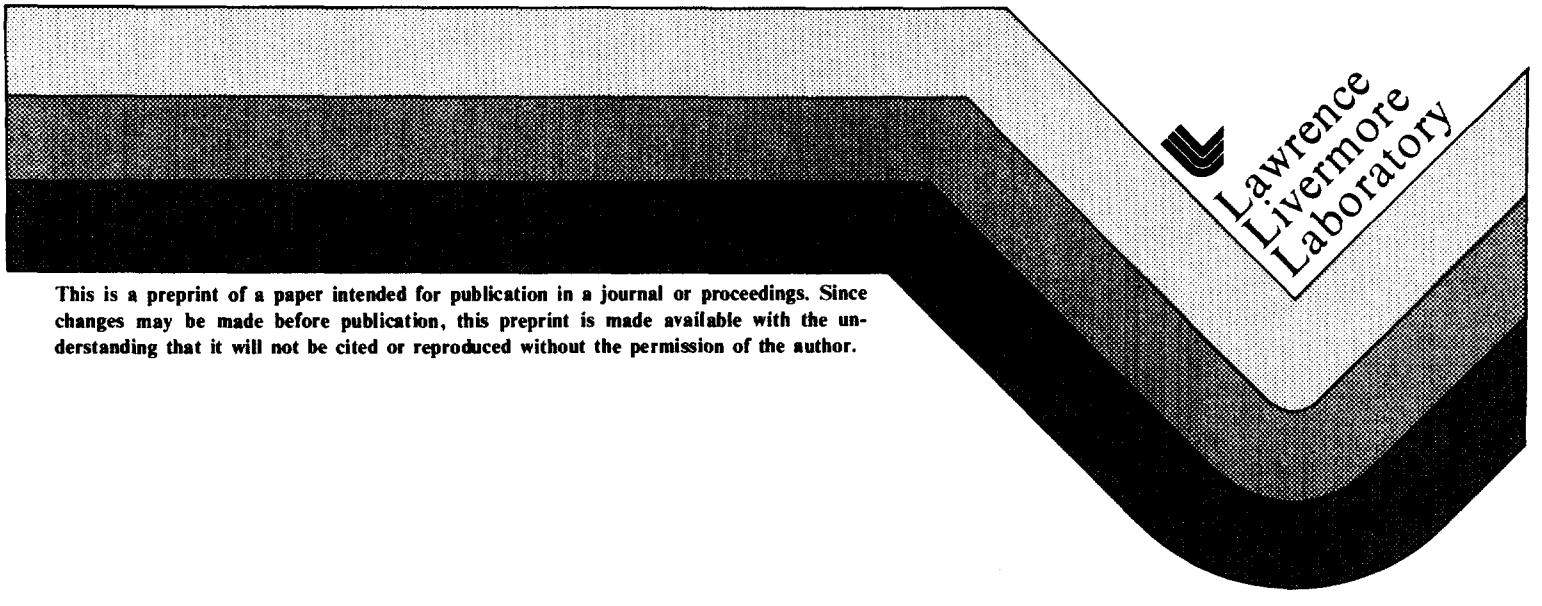

PRODUCTION AND LOSS OF H^- and D^- IN THE VOLUME OF A PLASMA

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ABSTRACT

The study of the production and loss of negative ions, H^- and D^-, in the volume of a plasma has received considerable attention since the measurement of anomalously high densities of H^- in 1977. The most probable mechanism for production is dissociative attachment (DA) to vibrationally highly-excited hydrogen molecules. New diagnostics developed for this purpose are photodetachment and the extension of coherent anti-Stokes Raman scattering (CARS) systems to the sensitivity required for low-pressure gases. Measurements and calculations indicate that the important loss mechanisms are diffusion to the walls at low densities and collisional destruction of several types at plasma densities above 10^{10} cm^{-3}. Production mechanisms must be highly efficient to compete with the losses. It appears to be straightforward to extrapolate measurements and theory to the densities above 10^{12} cm^{-3} that are required for an intense source of D^- for neutral beam injection into magnetically-confined fusion devices.

INTRODUCTION

For various reasons, the production and loss of negative hydrogen ions (H^-, D^-, etc.) in plasmas are now under study at several laboratories. The motivations for such investigations are both practical and fundamental. The diversity of motivations leads to interactions among several laboratories and several branches of physics.

Practical applications for negative ions are based on the fact that, after the ion is accelerated to high energy, it is easy to change the ion charge to produce either a neutral atom or a positive ion by detaching one or both of the electrons. It is less easy, on the other hand, to add an electron to the positive ion at energies above 100 keV. The electron detachment may be achieved by collisions with some target, which may be a thin foil, a cell filled with gas, a plasma, or photons. The negative ion may be produced either in the volume of a plasma, on surfaces, or in a gas or vapor cell.

At energies above 100 keV, a neutral atom can be produced more efficiently by electron detachment of negative ions than by collisions of positive ions. This fact leads to requirements for intense beams of negative ions for efficient injection of neutral atoms into magnetically-contained fusion devices such as tokamaks[1] or magnetic mirrors.[2]

Negative ions are useful in high-energy accelerators because after they are converted into positive ions they can easily be extracted from a cyclotron or accelerated a second time to attain an energy double the accelerator voltage. Such accelerator applications have now become routine.[3]

In addition to their practical applications, negative hydrogen ions are scientifically valuble because they are sufficiently simple; their properties can be precisely computed by theory. Theoretical principles thus proven can be applied to more complicated situations. Also, H^- ions are abundant in the outer layers of stars and our sun. They affect the emitted spectrum and can be used as observational tools.

PRODUCTION AND LOSS MECHANISMS

Production and loss of negative ions in plasmas is an interesting topic because of the many possible mechanisms that can be considered. A partial list is shown in Table I. Some of the loss mechanisms are extremely rapid. For example, $\overline{\sigma v} \approx 10^{-7}$ cm^3/s for mutual neutralization with positive ions.

Consequently, the production mechanisms must be extremely efficient if they are able to compete with the losses. It was discovered at Ecole Polytechnique a few years ago that the fraction of H^- measured in a plasma was 100 times larger than the fraction that could be computed from the

Table I. Mechanisms for the production of H^-.

- Dissociative attachment to H_2 or H_2 (v)
  $H_2 + e \rightarrow H_2^- \rightarrow H^- + H$
- Polar dissociation of H_2
  $H_2 + e \rightarrow H^- + H^+ + e$
- Dissociative recombination of H_3^+
  $H_3 + e \rightarrow H^- + H_2^+$ (v)
  (The electron may be captured from gas or wall molecules)

Mechanisms for the loss of H^-.

- Mutual neutralization with positive ions
  $H^- + (H^+ \text{ or } H_2^+ \text{ etc.}) \rightarrow H + [H \text{ or } H_2 \text{ (v)}]$
- Collisional electron detachment
  H^- + target ion or electron → H + e + target
- Wall neutralization
  (important at low density)
- Associative detachment
  (converse of dissociative attachment)

processes known at that time.[4] The anomalously high H^- concentration has stimulated work on the better understanding of the production mechanisms and also has stimulated the development of better plasma diagnostics to verify the measurements.

It was discovered recently that the cross section for H^- production by dissociative attachment (DA) to H_2, is a strong function of the excitation state that may be due to either vibrational, rotational, or electronic excitation.[5-7] This is due to the fact that the cross section for DA is a product of the cross section for electron attachment to the H_2 molecule and the probability that the H_2^- molecular ion will survive long enough to dissociate before the electron is re-emitted. The survival probability is a very strong function of the excitation state because the effective shape and size of the molecule depends on the internuclear separation. Cross sections for H^- or D^- production may therefore be enhanced by vibrational excitation by a factor as large as 10^6 or more compared with capture through the ground vibrational level.

Other possibilities for enhanced production of H^- have been considered. For example, the cross section for dissociative recombination of H_3^+ has recently been measured[8] as a maximum of 1.6×10^{-18} cm^2, which is not large enough to explain the high rate of H^- production. The metastable, electronically-excited state $H_2(^3\pi_u)$ has been considered as a source of H^-.[9] However, we expect that the lifetime of this excited state in a plasma is very short because of electron and wall collisions that convert the metastable $^3\pi_u$ state to the quickly-decaying $^3\Sigma_g$ state.

Table I lists the mechanisms for production and loss of H^- that are probably the most important in a plasma. This table indicates that H^- ions are derived from some intermediate specie such as vibrationally-excited molecules $H_2(v)$ or H_3^+. Therefore, it is necessary to consider the reaction rates for each specie involved if we wish to understand how the H^- production is affected by various plasma conditions. Consequently, we must also consider the processes listed in Tables II, III, and IV to understand the production and loss of $H_2(v)$, H_3^+, and H_2^+, all of which are derived from other species. Tables I through IV are not complete; the mechanisms listed are those believed to be the most important in this context.

We expect nonlinear behavior in the H^- production rate because of the sequential connection of these atomic reactions with each other. A group at Ecole Polytechnique has approached this complicated problem by measuring plasma compositions as conditions are changed and by correlating these measurements with the behavior expected from the atomic reaction rates.[10,11] The purpose of this paper is to recapitulate the results of this effort and to show to what extent it has been successful.

Most of the mechanisms listed in Tables I through IV are well known and require no special discussion. The E-V process proposed for production of molecules with very highly-excited states ($v > 6$) was proposed by W. Kunkel and analyzed theoretically by J. R. Hiskes.[12] The E-V process

Table II. Mechanisms for the production of vibrationally excited H_2 (v).

- Dissociative recombination of H_2^+ or H_3^+
  (see mechanisms for production of H^-)
- Mutual neutralization of H_2^+ and H_3^+
  (see mechanisms for loss of H^-)
- Proton transfer
  $H_2 + H_2^+ \rightarrow H + H_3$
- e-V collisions
  H_2 (v) + e (~1 eV) $\rightarrow$ H_2 (v + 1) + e
- E-V singlet collisions
  H_2 + e (~60 eV) $\rightarrow$ H_2^* (electronically excited)
  $\rightarrow$ H_2 (v) + hν

Mechanisms for the loss of vibrationally excited H_2 (v).

- V-T collisions with gas molecules
- Wall collisions
- Dissociative attachment
  (see mechanisms for production of H^-)

Table III. Mechanisms for the production and loss of H_3^+.

Production mechanism

- Proton transfer
  $H_2 + H_2^+ \rightarrow H + H_3^+$

Loss mechanisms

- Dissociative recombination
  (see mechanisms for production of H^-)
- Wall recombination
- Mutual neutralization

Table IV. Mechanisms for the production and loss of H_2^+.

Production mechanism

- Ionization of H_2 by electrons and ions

Loss mechanisms

- Wall neutralization
- Proton transfer
  (see mechanisms for production of H_3)

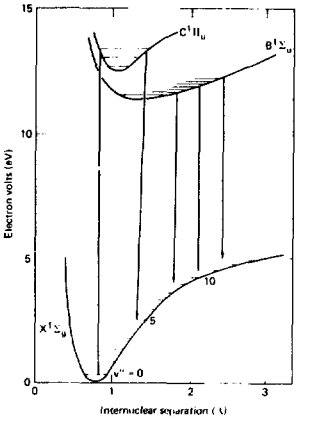

FIG. 1. The E-V process for producing excited molecules. An energetic electron excites a bound electron to a electronically-excited singlet state that initially has the same internuclear separation as the ground-state H_2. When the electronic excitation decays the internuclear separation has changed, producing a vibration-ally-excited H_2 (v).

mentioned in Table II for producing vibrationally-excited H_2(v) is illustrated in Fig. 1. This figure shows three states of an H_2 molecule as a function of the intermolecular separation.

The intermolecular separation will not instantaneously change as the ground-state H_2 is electronically excited. The electronically-excited state will probably find itself in a state of vibrational excit tion because the intermolecular separation does not correspond to its v = 0 state. The excited state will execute several vibrations during its excited lifetime. When it finally electronically decays, the intermolecular separation will not be the same as before; therefore, the molecule is electronically excited.

E-V collisions require an electron energy of 20 to 100 eV for the electronic excitation. On the other hand, e-V collisions for vibrational excitation can be caused by electrons of energy 1 eV or more. Therefore, the analysis must include both populations that produce vibrational excitation by the two methods. The E-V process is more effective for production of highly-excited vibrational states than the e-V process. However, only a small fraction of the electrons is sufficiently energetic to excite E-V collisions.

The vibrational lifetime is very long if there are no collisions. One of the unknown factors in Table II is the average number of wall collisions (b) required to de-excite the molecule.

THEORETICAL SCALING LAWS

If the only important H^- production mechanism is dissociative attachment to the vibrationally-excited H_2, the production rate in ions/cm^3 • s is

$$n_e n_v \overline{\sigma v}_{DA} \quad . \tag{1}$$

The density $n_v = b\,A\,n_+$ of vibrationally-excited H_2 is proportional to the plasma density n_+ and also is proportional to the parameter b (the average number of wall collisions the $H_2(v)$ can undergo before becoming de-excited).

The rate of loss of H^- by collisional electron detachment (ED), mutual neutralization (MN), and diffusion to the walls is

$$n_- n_e \overline{\sigma v}_{ED} + n_- n_+ \overline{\sigma v}_{MN} + n_-/\tau_d \quad , \tag{2}$$

where τ_d is the average time required for an H^- to diffuse out of the positive potential of the plasma. The plasma potential depends upon the electron temperature, and therefore, may vary with plasma density.

By equating the production rate, Eq. (1), and loss rate, Eq. (2) (using $n_v = b\,A\,n_+$), we obtain a scaling law

$$n_- = \frac{bA\,n_e^2\,\overline{\sigma v}_{DA}}{n_e\,\overline{\sigma v}_{CD} + 1/\tau_d} \quad , \tag{3}$$

where we have used, for simplicity, $n_e \approx n_+$ and have defined a total rate coefficient for collisional destruction of H^-:

$$\overline{\sigma v}_{CD} = \overline{\sigma v}_{ED} + \overline{\sigma v}_{MN} \tag{4}$$

The scaling law, Eq. (3), indicates two regimes depending upon which term in the denominator is dominant.

Regime (1)

At low density, the collisional destruction of H^- is negligible and Eq. (3) becomes

$$n_- = bA\,\tau_d n_e^2\,\overline{\sigma v}_{DA} \quad , \tag{5}$$

which indicates that n_- increases in proportion to n_e^2 or n_e^3, depending upon whether $\overline{\sigma v}_{DA}$ or τ_d increase with n_e.

Regime (2)

At high density, the diffusion loss is negligible and Eq. (3) becomes

$$n_- = bAn_e\,\overline{\sigma v}_{DA}/\overline{\sigma v}_{CD} \quad , \tag{6}$$

which indicates that n_- continues to increase linearly in proportion to n_e if $\overline{\sigma v}_{DA}$ and $\overline{\sigma v}_{CD}$ is constant, i.e., if the electron temperature does not vary with plasma density.

Therefore, if n_- continues to be plotted as a function of n_e on a log-log scale, we expect to find two linear regimes, indicated by Eqs. (5) and (6). The transition between the two regimes occurs when the two terms in the denominator of Eq. (3) are equal. The transition density is

$$n_e = (\tau_d\,\overline{\sigma v}_{CD})^{-1} \approx 10^{10}\ \text{cm}^{-3} \quad . \tag{7}$$

DIAGNOSTIC TECHNIQUES FOR H^- PLASMAS

Because anomalously high proportions of H^- were found in low density plasmas, there has been some recent motivation to improve diagnostics for plasmas containing H^-. One of the oldest and most useful techniques is the use of Langmuir probes to measure densities of positive ions and electrons in plasmas. Using the original Langmuir theory for a thin cylindrical probe, Doucet[13] proposed a method for measuring negative-ion density in a plasma. Using this method, Bacal et al[14] measured negative hydrogen-ion densities up to 30% of the positive-ion density.

If the probe characteristic curve is properly parabolic in shape, the positive-ion density n_+ can be determined from the ion current:

$$i_+ = n_+\,q\,d\ell\,(2q\,V_+/m_+)^{1/2} \quad . \tag{8}$$

For equal probe voltages ($V_+ = -V_-$) the ratio of electron current to positive-ion current is

$$i_e/i_+ = (n_e/n_+)\,(m_+/m_e)^{1/2} \tag{9}$$

Normally, the electron current i_e is much greater than the negativeion current i_- because of the difference in mass. Equation (9) requires

a correction term (not shown), if i_- is not negligible in comparison with i_e.

The negative-ion density n_- is

$$n_- = n_+(1 - n_e/n_+) \quad , \tag{10}$$

where n_+ and n_e/n_+ are obtained from Eqs. (8) and (9).

However, the probe method has some inaccuracies; it is also constrained by the limitations of the Langmuir theory. The Langmuir theory is usually useful only for low-density plasmas in which the probe radius is smaller than the Debye distance. It has been pointed out[15] that for different versions of probe theories, Eq. (8) changes by a factor of 1.52. Also the probe voltage V is defined relative to the plasma potential, which may not be definitely known. In one example a small change in the assumed plasma potential caused a 20% error in the measurement of n_-.[15] The measured n_- depends on the effective positive ion mass m_+ that, in turn, depends on the content of impurities and molecular ions. The probe technique is useful for gross measurements of n_+ and n_- but must be verified by other techniques.

A preferred technique for measuring the content of positive and negative ions in a plasma is by extraction and mass analysis.[4,10] This technique provides mass spectra of positive and negative ions. However, the extraction conditions sometimes change when the polarity of the extraction voltage is reversed. Therefore, it is not safe to measure the ratio n_-/n_+ by extraction and mass analysis.

A better technique for the measurement of n_- is by photodetachment, using an intense light pulse to detach the electron:

$$H^- + h\nu \rightarrow H + e \quad . \tag{11}$$

This method was proposed by Taillet[16] and developed for the case of a hydrogen plasma by Bacal et al.[14]

The fraction of the plasma negative ions destroyed by a light pulse is

$$\Delta n_-/n_- = 1 - \exp[-(\sigma/h\nu)(\text{laser pulse energy/area})]. \tag{12}$$

For each negative ion destroyed by photodetachment, an electron is produced. Therefore, we measure the increase in electron density after the light pulse with no measureable change in positive-ion density. The technique can be authenticated by changing the laser pulse energy and ascertaining that the measurement of $\Delta n = -\Delta n_e$ is consistent with Eq. (12).

The photon energy $h\nu$ should be chosen larger than the electron affinity of hydrogen (0.75 eV), but smaller than the electron affinities and ionization potentials of impurities. At Ecole Polytechnique we first used a ruby laser that had a photon energy of 1.8 eV. Recently a neodymium glass laser with a photon energy of 1.2 eV has replaced the ruby laser. In either case, the cross section for photodetachment of H^- is near the maximum of 4×10^{-17} cm^2.

Figure 2 shows how the measurements $\Delta n_-/n_-$ agree with Eq. (13) and thereby prove that the measurements are due to photodetachment of H^- and not of some other specie such as O^-, OH^-, or O_2^-. Cross sections for photodetachment of these other species are one or two orders of magnitude smaller because of the greater electron affinity. Photodetachment of OH^- and O^- is further excluded using the neodymium laser because of the low photon energy.

The change in electron density Δn_e can be measured by probes, interferometer, or other techniques. For measurement of the ratio $\Delta n_e/n_e$, it is not essential that the probe diameter satisfy the Langmuir theory. It is convenient for alignment to use a probe large enough to be rigid and self-supporting. Figure 3 shows the

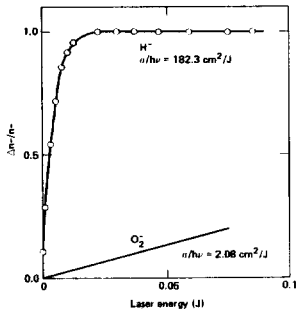

FIG. 2. Photodetachment of several negative-ion species by neodymium laser light as a function of laser pulse energy according to Eq. (12), for which $\sigma/h\nu = 182.3$ cm^2/J for H^- and 2.08 cm^2/J for O_2^- and the area of the laser beam is 0.71 cm^2.

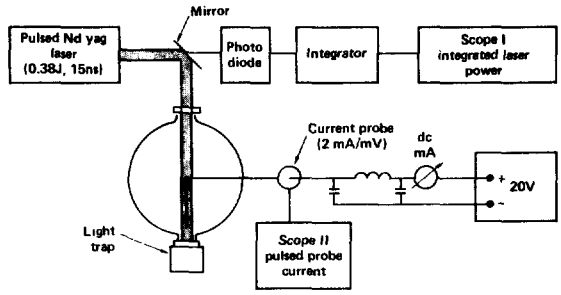

FIG. 3. Equipment diagram for measurement of H⁻ density by photodetachment.

photodetachment equipment diagram. Figure 4 illustrates how the probe was protected by a cylindrical screen to prevent the laser light from hitting the probe surface. The screen was necessary when ruby laser light of 1.8 eV photon energy was used, but not when neodymium laser light at 1.2 eV was used.

The photodetachment system shown in Fig. 3 was usually operated by applying a positive bias voltage and measuring the dc and pulsed electron currents collected by the probe. For laser pulse energies above 0.02 J, it was expected that the pulsed current Δi would be proportional to n_- and that the dc current i_{DC} would be proportional to n_e. To test this assumption the electron current collected by the large probe was compared with measurements of n_+ and n_e by the classical Langmuir probe as the gas pressure was changed. This test showed that the i_{DC} was more nearly in proportion to n_+ than n_e. The measured n_e increased by a factor of 1.7, while

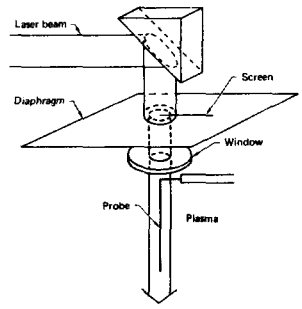

FIG. 4. Details of the equipment used in photodetachment measurements. The 0.5-mm probe diameter is sufficient for rigidity. A cylindrical screen is required to prevent the laser light from hitting the probe surface when using a ruby laser of 1.8 eV photon energy. It is not required when using a neodymium laser of 1.2 eV photon energy. The diaphragm limits the laser beam diameter.

i_{DC} and n_+ were maintained constant. This factor may represent the the inaccuracy of measuring n_e by a Langmuir probe.

The importance of vibrational excitation on the cross section for H^- production by DA has motivated the development of techniques for measuring the vibrational distribution. A standard technique for this measurement at high pressure is Coherent Anti-Stokes Raman Scattering (CARS), in which two colinear laser beams traverse an Raman-active medium such as vibrating molecules.[17] If the difference in frequency ($\Delta f = f_1 - f_2$) of the two light beams is equal to the molecular vibration frequency f_v, a new wave is generated at the anti-Stokes frequency

$$f_3 = f_1 + f_v = 2f_1 - f_2$$

This technique works best at high pressures; the instrumental sensitivity for a given state is about 10^{12} cm^{-3}.

PARAMETRIC STUDIES

To test the theoretical model, which includes the main atomic reactions and the scaling laws,[7] a series of parametric studies has been conducted at Ecole Polytechnique in low-density plasmas with substantial fractions of H^- and D^-. These measurements have been reported elsewhere.[4,7,10,11] We will only present a summary of the results at this time.

In the parametric studies the effects of plasma density, gas pressure, electron temperature, isotope, magnetic confinement, wall conditions, and impurities were examined. If possible, only one of these parameters was varied at a time. Changes in parameters were measured when they could not be experimentally controlled. The results were as follows:

a. Density dependence. In agreement with Eq. (5), n_- was measured at low density to increase in proportion with n_e^2 or n_e^3, depending upon conditions affecting the electron temperature.[7,10] The maximum ratio n_-/n_e was in the range of 20 to 35%. When the plasma density was increased above 10^{10} cm^{-3}, n_- continued to increase as shown in Fig. 5, but only in proportion to n_e, as predicted by Eq. (6).[11] For technical reasons, the maximum density n_- reported was 10^{10} cm^{-3} (not shown in Fig. 5), while n_e was 5×10^{10} cm^{-3}.

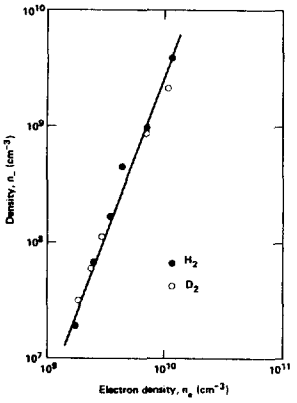

FIG. 5. Dependence of n_- on n_e in one example of low-pressure plasma. At about $n_e = 10^{10}$ cm^{-3}, the slope changes on a log-log scale in agreement with Eqs. (5) and (6).

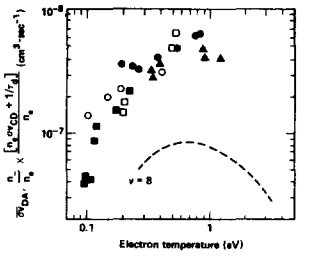

FIG. 6. Dependence of n_- on electron temperature, normalized according to the scaling law, Eq. (3). The dotted line is the theoretical $\overline{\sigma v}_{DA}$ for v = 8. Both the normalized n_- and $\overline{\sigma v}_{DA}$ have the same dependence on electron temperature.

The upward trend indicates that with technical improvements, n_- can be increased to the range above 10^{12} cm^{-3}. A data point on this extrapolation can be obtained from the performance of Ehlers' intense H^- source[18] which in 1965 emitted a volume-produced H^- beam of 33 mA cm^{-2}, which roughly indicates that $n_- \approx 5 \times 10^{11}$ cm^{-3}.

b. Gas pressure dependence. The scaling laws predict no dependence of n_- on gas pressure, except for indirect effects. It was established that within a pressure range from 0.4 to 4.0 x 10^{-2} Torr, the gas pressure had no effects other than those attributed to changes in electron temperature.

c. Electron temperature dependence. The scaling laws predict that the electron temperature affects the difussion time τ_d and also the rate coefficients $\overline{\sigma v}_{DA}$ and $\overline{\sigma v}_{ED}$. At low density, $\overline{\sigma v}_{ED}$ is not important. Figure 6 shows that, as a function of electron temperature, measurements indicated that the ratio $n_-(n_+ \overline{\sigma v}_{CD} + 1/\tau_d)n_e n_+$ was proportional to the theoretical rate coefficient $\overline{\sigma v}_{DA}$. This tends to confirm the scaling laws and also the belief that DA is the dominant production mechanism.

d. Isotope dependence. The photodetachment and mass-analysis measurements indicate no measureable difference between the production rates of H^- and D^-. This is consistent with the theory of dissociative attachment only if the H_2 or D_2 molecule is vibrationally excited to quantum states higher than v = 6. For the lower vibrational states, the cross section for DA is several orders of magnitude lower for D^- than for H^-. This indicates that the vibrational states higher than v = 6 are responsible for the high rate of D^- production. Bacal et al[10,11] estimated from these data that the average number of wall collisions b required for de-excitation was in the range from 6 to 19.

e. Wall dependence. Several effects were noticed when the inside wall of the plasma chamber was changed from glass to metal. The dependence of n_- on n_e changed from n_e^3 to n_e^2 at an intermediate gas pressure, but tended toward n_e^3 at the lowest gas pressure (3 x 10^{-3} Torr) at which operation was possible. Consequently, the maximum fraction n_-/n_e was reduced from 35% in the glass chamber to 15% in the metal chamber. Interpretation of these observations is difficult because several types of wall effects are possible.

f. Effect of magnetic confinement. Plasma confinement by an array of multipole magnets had the effect of increasing both the plasma density and n_-, and made operation possible at lower gas pressures. The plasma potential was reduced from 6.5 to 3 eV because the electrons were confined partially by the magnetic field.

g. Effect of impurities. In order to measure high concentrations of H^- or D^-, it was invariably found necessary to condense the impurities by a cold trap. We believe this is the result of the destruction of H^- by water vapor, which is known to have a high reaction rate.

$$H^- + H_2O \rightarrow OH^- + H_2 \qquad (13)$$

h. Measurements of vibrational excitation. For reasons of instrumental sensitivity, the measurements of vibrational excitation by CARS were conducted with a hydrogen gas pressure (>0.1 Torr) at least one order of magnitude higher than the gas pressure in which the H^- ion density was measured. Consequently, not all of the results were directly relevant to the low-pressure discharge. Measurements of the first three levels[19] indicate that about 2.5% of the molecules were vibrationally excited. The population of the state v = 1 was 40 times less than the population of the state v = 0; the population of the state v = 2 was 10 times less than that of the state v = 1. This indicates that the vibrational distribution is non-Boltzman. From these measurements, Hiskes concluded that the average number of wall collisions b required for de-excitation was in the range from 7 to 14.[20]

CONCLUSIONS

It appears clear that the high rate of production of D^- and H^- in plasmas can be explained by dissociative attachment to hydrogen molecules of vibrational quantum states higher than v = 6. The high excitation numbers are required to explain the measurements for two reasons: the greatly enhanced production rate and the lack of isotope effect, which is very large for low-lying excited states.

Highly excited vibrational states can be produced more effectively by E-V collisions than by e-V collisions, which are characterized by the transition $\overline{\sigma v} = \pm 1$. Molecular hydrogen ion dissociative recombination on walls is also imprortrant. At still higher densities reactions involving two charged particles become dominant. The production rates of $H_2(v)$ and of H^- are sufficiently efficient to compete with the losses even at high density.

Scaling laws agree with experimental measurements. These indicate that D^- densities can be scaled up to the range above 10^{12} cm^{-3}, which is required for intense sources of D^- for efficient neutral injection into large fusion devices such as tokamaks and magnetic mirrors.

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